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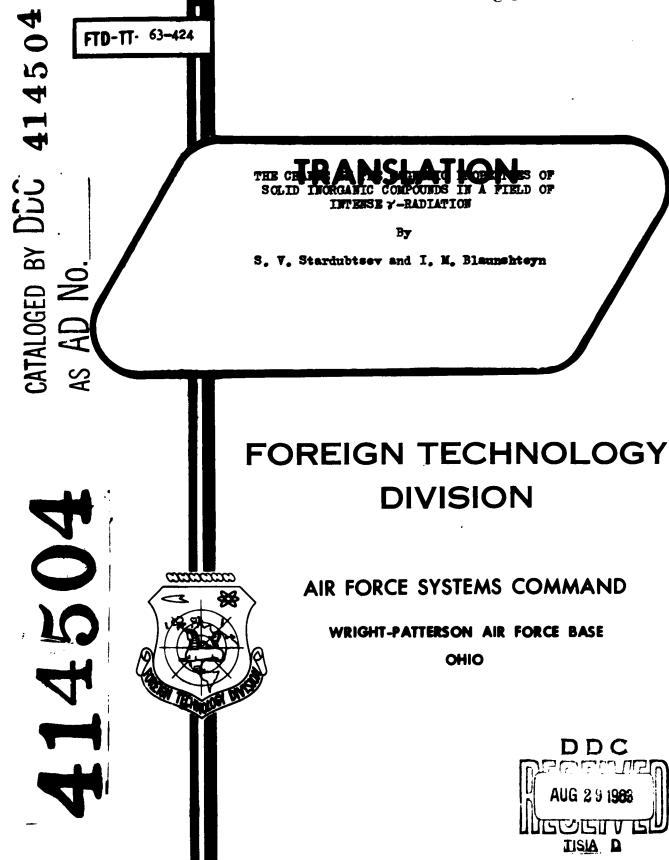
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# **UNEDITED ROUGH DRAFT TRANSLATION**

THE CHANGE IN THE MAGNETIC PROPERTIES OF SOLID INORGANIC COMPOUNDS IN A FIELD OF INTENSE  $\gamma$ -RADIATION

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## THE CHANGE IN THE MAGNETIC PROPERTIES OF SOLID INORGANIC COMPOUNDS IN A FIELD OF INTENSE $\gamma$ -RADIATION

### S. V. Starodubtsev and I. M. Blaunshteyn

Nuclear radiations acting on a substance cause a number of disturbances and conversions in it and change many of its properties.

Among the properties sensitive to radiation effects is a complex of magnetic properties. Therefore, it was of interest to us to study the changes in the magnetic properties of certain inorganic compounds subjected to  $\gamma$ -irradiation.

Ion, ion-covalent crystals, and certain elements with pronounced semiconductor properties were among the objects investigated.

The literature indicates that the diamagnetism of quartz, aluminum oxide, and magnesium oxide decreases during neutron irradiation [1]. This effect, in the opinion of the authors, is connected with the formation of paramagnetic centers representing in some cases interstitial atoms, in others - coloration centers, or simultaneously both one and the other.

In certain papers [2-5] a significant change in the hysteresis loop of certain ferrites was detected after irradiation. Although the theory of this question is far from clear, these effects are already

finding application in electronic-computer engineering and radio engineering.

The magnetic moment of a specimen is the result of the interaction between the spin and orbital angular momenta of all its atoms. In the majority of cases a solid has a structure such that the components of the magnetic moments are, as a rule, compensated, and this leads to diamagnetism. Irradiation first of all disturbs the electronic structure of the solid because of atomic and molecular ionization and excitation, which changes the total moment of the solid. In insulators and semiconductors, because of their low electrical conductivity, these changes may be retained for a fairly long time and can be measured by static methods. Irradiation most often induces a paramagnetic component in the susceptibility. In paramagnetic solids irradiation may redistribute the electrons, so that part of the system passes into the lowest magnetic state, which is revealed by a decrease in the paramagnetism.

In the general case the susceptibility of a solid is composed of three components:

$$x = \chi_i + \chi_i + \chi_e$$

where  $\chi_l$  is the resultant lattice moment, which varies mainly during irradiation by heavy particles;  $\chi_l$  is the moment of the current-carriers, which varies during irradiation by any radiation;  $\chi_c$  is the moment of the impurity-atoms, which is made to vary and is induced by any radiation, particularly in chemical compounds. Irradiation can influence any of these components, whence it follows that the susceptibility can be extremely sensitive to irradiation.

Change in Diamagnetism and Paramagnetism of Various Compounds when Irradiated

Irradiated material	Increase in Increase in diamagnetissparamagnet- (decrease inism (de- paramagnet- netism) diamagnet-		Type of magnetism	
		'ism)		
BaCi <sub>e</sub>		•	Diamagnetic	
אכנ	,	i -	•	
NaCl	i			
K.MnO.	: <del>-</del>	1	Paramagnetic	
KNO,	ļ <u>-</u> .		Diamagnetic	
NaNO,		j <i>→</i> -	1	
BaCO,	+	•	<b>.</b>	
Nico,	1 4-	<b>i</b>	Paramagnetic	
a.Fe.O.		+	Antiferromagnetic	
(:r <sub>2</sub> O <sub>3</sub>	1	<del> </del> +-		
FeCl,	İ	+	! <b>*</b>	
FeCl,	ļ	+++++++++++++++++++++++++++++++++++++++	>	
FeS		i +	•	
Co.O,	+		•	
CoCl,	-+-		•	
FeSO,			Paresagnetic	
BaO	<u> </u>		Dismagnetic	
CaO		•	! *	
MgO				
ZnO	i		,	
CuCl		••	•	
SiC	!	+	Paramagnetic	
CdSe	-		•	
Se	+		Diemagnetio	
Silice gel	+ + +		•	
Seignette salt	-		l ,	

In practice, the determination of  $\chi$  reduces most often to determining the force F acting on the specimen suspended in an inhomogeneous magnetic field. It is known that this force is expressed as follows:

$$F = \frac{1}{2} (\chi_1 - \chi_2) (H_1^2 - H_2^2) S$$

where  $\chi_1$  and  $\chi_2$  are the specific susceptibilities of the specimen and the ambient medium;  $H_1$  and  $H_2$  are the maximum and minimum field along the length of the specimen; S is the cross-sectional area of the specimen.

In our experiments the field was created by an electromagnet and amounted to 10,000 oersteds for a current of 10 amp. The geometry of FTD-TT-63-424/1+2+4

the field was recorded, in order to select the point of the field with a maximum gradient. The force was determined by a VA-200 analytic balance with an accuracy of 0.02 mg. Since we were interested in the change in the susceptibility under irradiation, we did not carry out absolute measurements of  $\chi$ .

The table reflects the qualitative character of the change in the magnetism of the objects which we irradiated. The irradiation was accomplished by  $Co^{60}$   $\gamma$ -rays at a dosage rate of 330 thousand r/hr. For convenience of examination we divided the investigated objects into groups. Thus, the ionic compounds BaCl2, KI, and NaCl do not show a change in  $\chi$  (if there is any change, it is less than the sensitivity of the method of determination). In order to ascertain the reasons for this fact, we investigated the gas liberation of these salts. was found that with the exception of BaCl2, which has a chlorine yield of only 0.003 mole per 100 ev, the remaining salts were not subjected to radiolysis (Fig. 1). Their coloration, as was established long ago, is related to the capture of  $\gamma$ -ray-ejected electrons by defects present in the crystal. The number of barium and chlorine atoms in BaCl2 formed during radiolysis and remaining in the specimen in the form of impurities is too small to introduce a noticeable contribution to the magnetism of the basic salt. Their effect on the electronic structure of the conductance is also, obviously, insignificant, since BaCl2 is not a semiconductor. It is curious to notice the absence of  $\Delta \chi$  in KMnO4, although the fact that it undergoes strong decomposition during  $\gamma$ -radiation is established (Fig. 1).

In sodium and potassium nitrates the diamagnetic susceptibility is decreased, and in the case of a dose of 40 million <u>r</u> the changes are 12 and 20% for NaNo<sub>3</sub> and KNO<sub>3</sub>, respectively. Figure 2 shows these changes. The relative change in the force acting on a specimen in a

magnetic field before ( $\Delta p$ ) and after ( $\Delta p$ ) irradiation is plotted along the y-axis. It is known [6] that these nitrates noticeably decopose in a field of  $\gamma$ -rays and are converted into nitrites, with oxygen gas being liberated and neutral atoms of sodium and potassium being formed. It is possible that the relative change of  $\chi$  in KNOs is more than in NaNOs due to the larger yield and the larger difference  $\chi_{1\text{on}} - \chi_{\text{atom}}$  in the case of potassium than in the case of sodium.

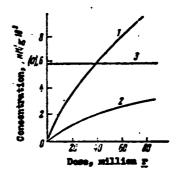


Fig. 1. Gas liberation during irradiation of ionic compounds: 1) KMnO4; 2) BaCl<sub>2</sub>; for KI and NaCl the origin of the coordinates is moved to a point with the ordinate 6.

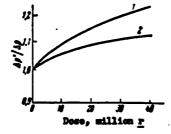


Fig. 2. Relative change in the force acting on a nitrate specimen in a magnetic field versus dose: 1) KNO<sub>3</sub>; 2) NaNO<sub>3</sub>.

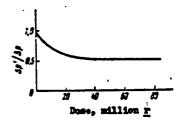


Fig. 3. Relative change in the force acting on a CuCl specimen in a magnetic field versus dose.

A special group of investigated compounds includes a-Fe<sub>2</sub>O<sub>3</sub>, Cr<sub>2</sub>O<sub>3</sub>, FeCl2, FeCl3, FeS, Co2O3, CoCl2, which, from the magnetic point of view, are antiferromagnetics. The change in their susceptibility sometimes reaches 50% (Co2O3), although experiments on gas liberation indicated their relative radiation stability. We can find an explanation for this fact by using the contemporary theory of antiferromagnetism, according to which the components of the magnetic moments of the sublattices of oxides and chlorides have different magnetization directions. If the sublattices have an identical moment of absolute magnetization, then the total moment of the crystal is equal to zero (compensated antiferromagnetism); if, on the other hand, the sublattices have different moments, then the resultant moment is not equal to zero (noncompensated antiferromagnetism). There is a paper [7] in which it is pointed out that natural specimens of  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub>, together with intrinsic antiferromagnetism, have a small "parasitic" ferromagnetism in a direction parallel to one of the sublattices. This, in the opinion of the authors, is due to lattice defects. If this is really so, then those insignificant interstitial atoms which may appear as a result of radiolysis and remain inside will act in the direction of decompensation of the moment, i.e., they will increase the magnetic moment, which was observed by us in the case of a-Fe<sub>2</sub>O<sub>3</sub>, Cr<sub>2</sub>O<sub>3</sub>, FeCl<sub>2</sub>, FeS.

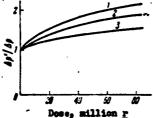


Fig. 4. Relative change in the force acting on an oxide sample in a magnetic field versus dose:
1) ZnO; 2) MgO;
3) BaO.

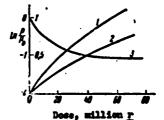


Fig. 5. Dependence of radiolysis of metal oxides on dose:
1) MgO; 2) BaO; 3) ZnO.

On the other hand, in the case of Co<sub>2</sub>O<sub>3</sub> and CoCl<sub>3</sub> the paramagnetism is decreased, which can be explained by Cramers' and Andersen's theory of "indirect exchange" [8], according to which an antiferromagnetic interaction is brought about between the elementary magnets (the moments of the cations of one of the sublattices) by means of the anions O and Cl , which are devoid of any moment. From the authors' investigation of the behavior of CaO it follows that unexcited oxygenions cannot lead to indirect exchange.

It is possible that during irradiation unexcited oxygen and chlorine atoms really do appear and that they promote a greater magnetic interaction and lead to a greater compensation of the magnetization of the Co<sub>2</sub>O<sub>3</sub> and CoCl<sub>2</sub> sublattices. Incidentally, the life of atom in an excited state is limited, which agrees with the experimentally observed annealing of the susceptibility.

We investigated CuCl in detail. After irradiation its diamagnetism is decreased with a transition in certain specimens to predominate paramagnetism. Figure 3 shows the relative changes in susceptibility versus the irradiation dose. Vacuum specimens do not show susceptibility changes. We explain this fact by the formation of paramagnetic centers in the form of CuCl<sub>2</sub>, or CuO and CuOCl. This explanation justifies itself somewhat by the fact that irradiated CuCl does not liberate chlorine gas.

We can include in another group the metallic oxides CaO, MgO, BaO, and ZnO, whose diamagnetic moment increases during irradiation. This is illustrated by Fig. 4. The greatest changes were observed in ZnO.

Tests on gas liberation showed that CaO, MgO, and BaO dissociate and liberate oxygen gas. On the other hand, ZnO, when irradiated, acquires adsorption properties (Fig. 5). We shall assume that the cause of the susceptibility change is the oxygen impurity formed during radiolysis.

This is confirmed to some extent by the fact that vacuum specimens change their susceptibility very little, since a vacuum furthers the output of gas beyond the limits of the specimen. In the case under consideration an oxygen impurity highly affects the electronic and energy structure of these semiconductors. Otherwise, a decrease in the diamagnetism would have been observed on account of the additive contribution of the paramagnetism of the impurity atoms of oxygen.

Note that when MgO is irradiated the diamagnetism decreases, which once again indicates the difference in the nature of the radiation disturbances when irradiated by light and by heavy radiation.

And, finally, such compounds and elements with semiconductor properties as CdSe and Se increase the diamagnetism when irradiated (Fig. 6 for CdSe). Figure 7 shows the time-dependent annealing for CdSe at room temperature and is of complex nature.

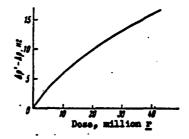


Fig. 6. Change in the force acting on a specimen of CdSe in a magnetic field versus dose.

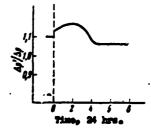


Fig. 7. Time-dependent annealing of a change occurring during irradiation of a specimen of CdSe at room temperature.

### CONCLUSIONS

- 1. The magnetic susceptibility of many inorganic compounds changes when irradiated by  $\gamma$ -rays.
- 2. The data obtained in this work are of qualitative nature and do not claim to be a complete and unique interpretation.

3. A study of the change in the susceptibility of irradiated objects yields additional knowledge about the character of radiational disturbances.

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